arXiv:cond-mat/0202300v3 [cond-mat.dis-nn] 29 Sep 2002

Order-Parameter Criticality of the d=3 Random-Field Ising Antiferromagnet $Fe_{0.85}Zn_{0.15}F_2$

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(Dated: February 1, 2008)

The critical exponent $\beta = 0.16 \pm 0.02$ for the random-field Ising model order parameter is determined using extinction-free magnetic x-ray scattering for $Fe_{0.85}Zn_{0.15}F_2$ in magnetic fields of 10 and 11 T. The observed value is consistent with other experimental random-field critical exponents, but disagrees sharply with Monte Carlo and exact ground state calculations on finite-sized systems.

PACS numbers: 61.10.Nz, 75.40.Cx, 75.50.Ee, 75.50.Lk

The Ising model is perhaps the most important model in statistical physics, with many applications that go well beyond the realm of physics. In 1944, the pure two-dimensional ($d = 2$) model was solved exactly by Onsager [1]. For the pure $d = 3$ Ising model, although various calculations and computer simulation techniques have proven extremely useful, no exact results are available. Nevertheless, there is extremely good agreement among theory, simulations, and experiments [2]. Hence, the model can be considered well understood. Through the years, the intellectual value of the Ising model has grown, particularly as a model of disorder. One of the most important of these models of disorder occurs when a random field is imposed which couples directly to the order parameter of the system. The most studied realization of this random-field Ising model (RFIM) is the diluted antiferromagnet in an applied magnetic field. Unlike the pure $d = 3$ Ising model, there is, so far, poor agreement between theory and simulations on the one hand and experiments on the other [2]. This has motivated us to measure the critical behavior of the staggered magnetization, the antiferromagnetic order parameter

$$M_s = M_0 t^\beta \tag{1}$$

where $t = (T_c(H) - T)/T_c(H)$, of the dilute Ising antiferromagnet in an applied field, since this is one of the most valuable yet also least characterized aspects of the experimental system. Our result for the order parameter exponent β provides an important quantitative experimental contribution toward a comprehensive understanding of the RFIM.

The dilute, insulating antiferromagnet $Fe_xZn_{1-x}F_2$ has a large single-ion anisotropy and is an extensively studied [2] $d = 3$ RFIM realization [3, 4]. Nevertheless, prior attempts to determine the critical behavior of the order parameter have been unsuccessful. This may be surprising since, in principle, one only needs to measure the temperature dependence of the Bragg scattering intensity, I_B, which is proportional to $(M_s)^2$, with a magnetic field H applied along the c-axis, the spin ordering direction. For two reasons, such measurements have proven very difficult in practice. First, neutron scattering on these high-quality crystals suffers greatly from the effects of extinction; the beam, upon transmission through the crystal, is depleted of neutrons satisfying the Bragg condition, resulting in the saturation of the measured value of I_B. Second, for magnetic concentrations x below the vacancy percolation threshold [5, 6], $x_v = 0.754$, domain formation obscures the RFIM critical behavior below the transition at $T_c(H)$ in $Fe_xZn_{1-x}F_2$ and its less anisotropic isomorph $Mn_xZn_{1-x}F_2$. Although the domains may be internally well ordered, I_B will be greatly diminished if the characteristic length scale for the domain structure is smaller than that of the spectrometer resolution; we will refer to this as micro-domain structure and it has been studied extensively in previous works [7]. Under severe extinction conditions, domain structure may relieve extinction and actually cause I_B to increase. Whether domain structure forms or not, the Bragg scattering cross section will be decreased by thermal disorder as the transition is approached. In x-ray scattering, since the magnetic scattering cross section is relatively small, the scattering intensity, obtained in a reflection geometry, does not suffer from extinction, as extensively discussed previously [8, 9]. The use of extinction-free magnetic x-ray scattering, and of a crystal with $x > x_v$ to avoid micro-domains, has allowed us to accurately characterize the order parameter critical behavior in $Fe_{0.85}Zn_{0.15}F_2$.

The magnetic x-ray scattering technique was employed for MnF_2 for $H = 0$ by Goldman *et al.* [8], and was then applied to $Mn_xZn_{1-x}F_2$ with $H > 0$ by Hill *et al.* [9]. Whereas the $H = 0$ study yielded the exponent β consistent with the $d = 3$ Ising model, the latter did not reveal the universal RFIM behavior [9], which would be consistent with $x < x_v$; the $H = 0$ transition temperature [10] and its field dependence [11] are consistent with

the concentration being very close to or below x_v. That the transition in Ref. [9] was obscured by micro-domains [5] is suggested by the zero slope of ${M_s}^2$ versus T as $T \to T_c(H)$. The present magnetic x-ray scattering measurements use $Fe_{0.85}Zn_{0.15}F_2$, for which x is well above x_v.

The measurements were made at the new high-field magnet facility on beam line 7-2 of the Stanford Synchrotron Radiation Laboratory. A monochromatic x-ray beam was obtained from the wiggler spectrum via a Si(111) double-crystal monochromator. X-ray energies between 14 and 13.5 keV were used, which resulted in a penetration depth of about 60 μm. The energy was tuned to minimize energy-sensitive multiple scattering [9]. The sample had a finely polished face, a few mm^2 in area, with the a-axis perpendicular to the polished face and the c-axis along the vertical field. The temperature of the crystal, mounted in a He atmosphere, was stable to approximately 10 mK. The transition temperature for $H = 0$ was measured to be $T_N = 66.7$ K, consistent with birefringence measurements on the same sample [12] and with a concentration $x = 0.85$ [10]. It has been shown that the antiferromagnetic transition at this magnetic concentration is stable at fields as high as $H = 18$ T [6]. For $H = 10$ and 11 T, the transitions are at $T_c = 64.2$ and 64.0 K, respectively. The lattice constants of the sample were determined to be approximately $a = 4.68$ Å and $c = 3.27$ Å near the transition temperature. The half-widths-at-half-maximum for the Bragg peaks were 4×10^{-4}, 4×10^{-3}, and 4×10^{-3} reciprocal lattice units for the transverse, longitudinal and vertical directions, respectively, at the (100) magnetic Bragg point, about which transverse $H = 0$ and $H = 11$ T and longitudinal $H = 10$ T scans were obtained. The sample was remounted between measurements at different fields, and therefore we normalized intensities using scans at $T = 47$ K. Three conventional thermal-cycling procedures were employed. In ZFC, the sample is cooled in zero field below $T_c(H)$, the field is applied, and the sample is warmed through $T_c(H)$, waiting at each temperature at least 20 min before taking data to let the temperature and system stabilize. In FC, the sample is cooled through $T_c(H)$ in the field, taking data as in ZFC. Field-heated (FH) data were taken by heating in the field after FC. The scans typically consisted of 41 points, about 15 of which covered the Bragg peak. At each point, the intensity was counted for 30 to 45 seconds, depending on the temperature of the scan.

Figure 1 shows the Bragg intensity for $H = 0$ and 10 T versus temperature, with the q and T independent backgrounds subtracted, where q is the distance in reciprocal space from the (100) antiferromagnetic Bragg point. The background depends on the precise experimental configuration, but not on the thermal cycling used to collect data, and is mostly from sources other than the crystal itself. For comparison of the background to the Bragg

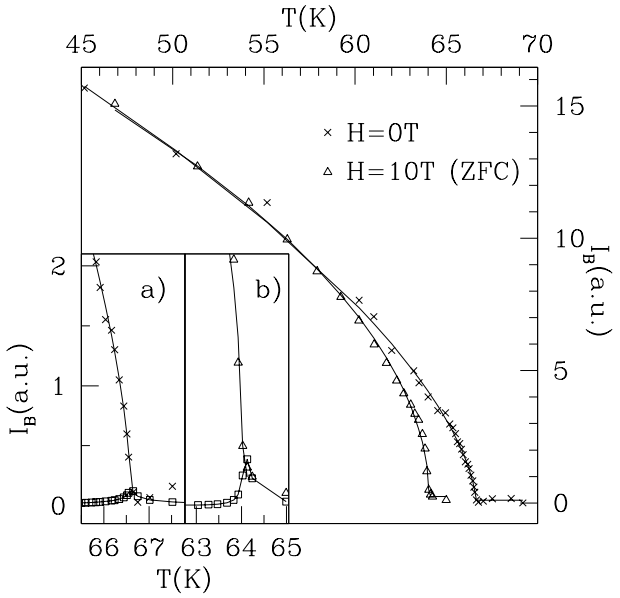

FIG. 1: The ZFC Bragg intensity, I_B, in arbitrary units (a.u.), versus T for $H = 0$ and $H = 10$ T, with the T-independent background intensity subtracted. The square symbols in insets a) and b) show the critical scattering contributions to the x-ray intensities for $H = 0$ and $H = 10$ T, respectively, determined from neutron scattering measurements as described in the text.

signal, typical background counts for the $H = 11$ T scans were eight counts per second whereas the $q = 0$ intensity was 160 counts per second at $T = 47$ K. Above the transition, the scattering intensity results only from the critical scattering and goes to zero well above $T_c(H)$, indicating that there are no contributions from multiple scattering to the Bragg intensity. To determine the critical scattering for $H = 0$, 10 and 11 T, neutron scattering line shapes, obtained with a sample of nearly the same magnetic concentration [13] using a previously described procedure [14], were folded with the x-ray resolution, and the overall $q = 0$ amplitude was adjusted to fit the $H = 10$ T data above $T_c(H)$. Insets a) and b) in Fig. 1 show the critical scattering contributions for $H = 0$ and 10 T, respectively. As a result of the high momentum resolution of the x-ray technique, the critical scattering contributions, which are nearly Lorentzian for $H = 0$ [14, 15], are almost negligible (Fig. 1a)). For $H > 0$, however, the critical scattering has a much larger q dependence [14] at small q. Consequently, a small contribution to the $q = 0$ scattering is more discernible for the $H = 10$ T data. These contributions were subtracted from all the data before determining the order parameter exponent.

Although neutron scattering measurements using $Fe_{0.85}Zn_{0.15}F_2$ [13] and $Fe_{0.93}Zn_{0.07}F_2$ [14] show no evidence for micro-domain formation in the critical scatter-

ing, $H > 0$ hysteresis in I_B is evident, with the FC intensities larger than the ZFC ones, a result of extinction. The x-ray Bragg scattering also shows hysteresis, but in this extinction-free case the ZFC data are higher in intensity. In the random field region, the FC intensity is factor of 4.1 smaller than ZFC intensity. This ratio depends slightly on the cooling rate used in obtaining the FC data. The ZFC data are rate independent. For $H = 11$ T, the corresponding ratio is approximately 4.0. FH data were intermediate between the ZFC and FC curves. We note that specific heat measurements also show hysteresis very close to $T_c(H)$ at this concentration [12, 16]. Such hysteresis is likely a result of the extremely slow activated RFIM dynamics and possibly represents the fact that FC long-range ordering must take place while traversing the transition, where activated dynamics plays the greatest role. The logarithmically slow relaxation associated with activated dynamics [17, 18] severely limits the ability of the system to equilibrate extremely close to $T_c(H)$. This limits the formation of long-range order upon FC. ZFC data, on the other hand, are obtained without approaching $T_c(H)$ except when the order parameter is already very small, and thus do not visibly suffer from the slow dynamics. Moreover, no dependence on the rate of temperature change was observed upon ZFC. Hence, we believe the ZFC data represent the correct order parameter measurement. Various measurements near the transition at this concentration have yielded critical behavior indicative of a second-order phase transition. There is also no measureable latent-heat in specific heat critical behavior measurements [12]. Therefore, this appears to be a second-order transition, although an extraordinary one.

The normalized Bragg intensity curves in Fig. 1 clearly approach $T_c(H)$ vertically. This is characteristic of experiments [13, 14, 15] and simulations [5] for $x > x_v$ and in stark contrast with experiments [2, 9, 19] and simulations [5] for $x < x_v$, where I_B approaches $T_c(H)$ horizontally. The latter behavior is attributable to micro-domain formation, which is energetically favorable when the vacancies percolate through the crystal, as shown in Monte Carlo simulations [5].

Figure 2 shows the logarithm of I_B, with the constant background and critical scattering contributions subtracted, for $H = 0$, 10, and 11 T, versus the logarithm of t. The values of $T_c(H)$ were determined from fits to the data. For $0.0007 < t < 0.03$ and $H = 0$, we find $\beta = 0.35 \pm 0.02$ (solid line), which agrees well with several experimental and theoretical determinations for the random-exchange Ising model [2]. For $H = 10$ and 11 T, a crossover from random-exchange to RFIM critical behavior occurs near $t = 0.03$, consistent with birefringence measurements [12], and the data can be fit to a single power law only in the range $0.0001 < t < 0.03$. The fits over this range yield the exponent $\beta = 0.16 \pm 0.02$ for the combined $H = 10$ and 11 T data and are indicated by the parallel solid lines in Fig. 2. A less sophisticated data

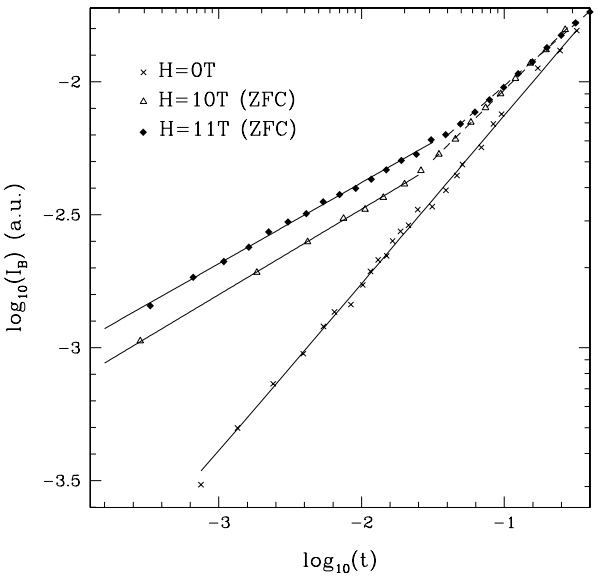

FIG. 2: The same ZFC data as in Fig. 1 as well as data taken at $H = 11$ T, corrected for the critical scattering contribution, plotted as the logarithm of the intensity versus the logarithm of t. The solid lines for $H = 10$ and 11 T indicate RFIM behavior with $\beta = 0.16$, while the solid line for $H = 0$ reflects conventional random-exchange behavior ($\beta = 0.35$).

analysis, which did not correct for the critical scattering contribution, resulted in a value for the order parameter exponent that is larger by 0.02, still within the error bars. The correction should, of course, be done in order to obtain the correct value of β. The slope of the data at large t changes with H since the definition of the reduced temperature involves $T_c(H)$ for $H > 0$ and not the $H = 0$ transition temperature T_N.

Through the Rushbrooke scaling relation

$$2\beta + \gamma + \alpha \geq 2 \quad , \tag{2}$$

which is usually satisfied as an equality, β is related to the universal critical exponents α (for the specific heat) and γ (for the staggered susceptibility) of the $d = 3$ RFIM. The experimentally determined specific heat peak is nearly logarithmic and very symmetric close to $T_c(H)$, consistent with $\alpha \approx 0$ [12, 16]. Neutron scattering analyses [13, 14] yield values in the $1.45 < \gamma < 1.65$ range. Therefore, the experimental value $\beta \approx 0.16$ is fairly consistent with Rushbrooke scaling, taking the upper limit of γ and $\alpha = 0$.

A very recent NMR study [20] of the order parameter in the effective short-range interaction random-field ferroelectric $Sr_{0.61-x}Ce_xBa_{0.39}Nb_2O_6$, with $x = 0.0066$, yielded $\beta = 0.14 \pm 0.03$, consistent with the present result for $Fe_{0.85}Zn_{0.15}F_2$. It was obtained, however, in the presence of micro-domain structure. Apparently, the more

local NMR probe is less sensitive than the Bragg scattering techniques to the formation of micro-domains. This suggests that domain formation does not preclude a fairly sharp RFIM-like phase transition and a measurement of its order parameter [21], but only prevents measurements of the order parameter through scattering experiments for $x < x_v$.

There does not exist a set of theoretical results that are consistent with all the experiments [2]. Monte Carlo [22] and exact ground state calculations [23] yield very small values for β and large, negative values for α. Other numerical and scaling analyses [24] yield α close to zero, consistent with experiments [12, 16], but also yield $\nu = 1.37 \pm 0.09$, much larger than experimental value $\nu = 1.05 \pm 0.01$ [13, 14]. Another recent work [25] yields α and β close to zero. One Monte Carlo study, on a large lattice and with less assurance of equilibrium than other simulations, yielded $\beta = 0.25 \pm 0.03$ [26]. Since consistency among numerical and experimental exponents continues to elude us, a comprehensive understanding of the $d = 3$ RFIM is yet to be achieved. The determination of the order parameter exponent presented here is an important quantitative contribution in this direction.

We thank M. Matsuda, S. Katano, H. Yoshizawa and J. A. Fernandez-Baca for allowing the use of unpublished neutron scattering results in the analysis shown in Fig. 1, and the SSRL staff for their help in building the magnet facility. The x-ray experiments were carried out at the Stanford Synchrotron Radiation Laboratory, a national user facility operated by Stanford University on behalf of the U.S. Department of Energy, Office of Basic Energy Sciences. The work at Stanford was also supported by the U.S. Department of Energy under Contract Nos. DE-FG03-99ER45773 and DE-AC03-76SF00515, by NSF Grant Nos. DMR-9985067 and DMR-9802737, and by the A.P. Sloan Foundation. The work at UCSC was supported by Department of Energy Grant No. DE-FG03-87ER45324. The work at West Virginia University was supported by NSF Grant No. DMR-9734051.

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